

MONITORED PLUTONIUM AEROSOLS AT A SOIL CLEANUP SITE ON JOHNSTON ATOLL

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MONITORED PLUTONIUM AEROSOLS AT A SOIL CLEANUP SITE ON JOHNSTON ATOLL.

Abstract: *Suspended plutonium in air was monitored for four periods near the operation of a stationary sorting system used to "mine" contaminated soil on Johnston Atoll. The monitoring periods were 14 Oct-14 Nov 1992, 20 Oct-15 Nov 1993, 16 Aug-3 Nov 1994, and 17 Feb-27 Feb 1995. Pairs of high volume air samplers were located at each of four locations of the process stream: the "spoils pile" that was the feedstock, the "plant area" near the hot soil gate of the sorter, the "clean pile" conveyer area where sorted clean soil was moved, and the "oversize soil" crushing area. These locations were monitored only during the working hours, while air monitoring was also done at an upwind, "background" area 24-hours per day. The four monitoring locations were extremely dusty during 1993-1995, with median total suspended particulate mass loading of 1070 $\mu\text{g}/\text{m}^3$ at the spoils pile, and maximum values of 5340 $\mu\text{g}/\text{m}^3$ at the oversize soil site during this period, when background mass loadings were typically 23 $\mu\text{g}/\text{m}^3$. Activity concentration of suspended Pu was 3.6 pCi/g in 1992, decreased to 0.43 pCi/g in 1993, and increased to 57 and 38 pCi/g in 1994 and 1995, respectively. This variation in activity concentration reflected the changing activity in the material being processed. The median concentrations of Pu in "workplace" air (combined spoils pile, plant area, and clean pile sites) in 1992 was 397 aCi/m³ (15 $\mu\text{Bq}/\text{m}^3$), but increased to median values of 23000 aCi/m³ (852 $\mu\text{Bq}/\text{m}^3$) in Aug-Nov 1994 and 29800 aCi/m³ (1100 $\mu\text{Bq}/\text{m}^3$) in Feb 1995. The highest median value at the work sites (29800 aCi/m³) was more than 200 times lower than the regulatory level. The highest observed value was 84200 aCi/m³ at the spoils pile site, and this was more than 70 times lower than the regulatory level. The conclusion was that, in spite of the dusty environment, and the increased level of specific activity, we did not find that the soil processing posed any significant risk to workers during the observation periods 1992-1995.*

INTRODUCTION

We monitored the air concentrations for suspended Pu-aerosol ($^{239,240}\text{Pu}$) during four separate periods of soil cleanup operation at Johnston Atoll from October 1992 to February 1995. The soil cleanup operation consisted of sorting the soil contaminated by plutonium (Pu) at Johnston Atoll, to produce soil clean enough for use in landfills while removing the high-activity, small soil volumes that are then disposed as waste (Bramlitt, 1993). This report provides results of the measurements of total suspended particulates in mass-loading ($\mu\text{g}/\text{m}^3$), mass-specific Pu-aerosol activity concentration (pCi/g), and Pu concentration in air (aCi/m^3). A previous study in 1992 by us (Shinn, Fry, and Johnson, 1994) provides additional details, such as particle-size and enhancement factor data.

METHODS

Air Sampling

Two high-volume air samplers (HV) were operated as a set at each of four separate locations during the working hours near the stationary soil-sorting system. Also, two "background" HV were operated continuously, together, at an upwind location, away from the soil contamination site. The HV were powered by a motor-blower unit mounted on a filter holder (General Metal Works, Inc., Model FH-2100), and cellulose fiber filters (8×10 -inch, Whatman 41) were used. This filter material is qualified by the American National Standards Institute, ANSI Standard N13.1 (reaffirmed 1993). At the face velocities in this application, for a test aerosol at $0.3 \mu\text{m}$ aerodynamic diameter Whatman 41 has 70% efficiency for fragmenting liquid aerosols and 92-94% efficiency for solid aerosols. For larger particles, it has efficiencies greater than 98%. Cellulose fiber is preferred to reduce the effort required for acid dissolution after filter exposure, but to measure mass loading a special effort was required to weigh the filters (after equilibration at a reference temperature and humidity) because of the hygroscopic nature of the media. The motor-blowers (General Metal Works, Inc) were operated for periods up to one month at a flow rate of $34 \text{ m}^3/\text{h}$. Flow rates and lapsed times were recorded daily, and flow rates were maintained by adjusting a variable autotransformer on each motor (VARIAC, Model 3PN116B, Superior Electric Co.) Flow rates were monitored at a pressure tap on the motor-blower using a dial manometer (Magnehelic Model 2002C). Calibration of the flow rates was performed in the laboratory using a transfer-standard orifice assembly (General Metal Works, Inc., Model GMW-25), which provided a linear

curve of flow rate versus the square root of pressure. Each motor was checked in the laboratory to see that it was leak-free and delivered a flow within 5% of the calibration for 34 m³/h at sea level. The HV were installed on portable tripod assemblies (General Metal Works, Inc., Model GT2200) and operated approximately 8 h per day during the working period. An operator covered the units with plastic bags for the periods when they were on standby. Details of HV air-sampler operating procedure are provided in "Protocols of Radiocontaminant Air Monitoring for Inhalation Exposure Estimates", (Shinn, 1995).

Four monitoring periods during operations of the cleanup (soil sorting) activities were: (1) 14 Oct to 14 Nov 1992, (2) 20 Oct to 15 Nov 1993, (3) 16 Aug to 3 Nov 1994, and (4) 17 Feb to 27 Feb 1995. The first period was reported earlier (Shinn, Fry, and Johnson, 1994). The third period was interrupted by a hurricane so that the sampling period was 16 Aug to 23 Aug and restarted, 30 Oct to 3 Nov 1994. (The air samplers were only operated when the sorting system was working.)

The "spoils pile" (SP) set of HV units was located near the feedstock where the contaminated soil was initially dumped onto a screen. The "plant area" (PA) set of HV units was located at the sorter hot soil gate near where containers were filled with the high Pu-activity soil. The "clean pile" (CP) set of HV units was located near the base of the conveyer unit that moved the sorted soil after it had passed successfully through the detector units. See the description of this facility for the "mining" of Pu provided by Bramlitt (1993). In addition, the "oversize soil" (OS) set of HV units were operated near a rock crusher that was used intermittently. The "background" HV units were operated continuously, 24 h per day for each period, and were located upwind where no Pu soil contamination could be found. The background HV filters were changed at the same time as other air samplers.

Radionuclide Analysis

Radiochemical methods were utilized to determine the ^{239,240}Pu, ²³⁸Pu, and ²⁴¹Am on aerosol particles impacted on the cellulose fiber substrate. Our minimum detectability limit was less than 0.3 pCi (0.01 Bq) per sample for these transuranic isotopes. The isotopes were recovered by acid total-dissolution, ion-exchange separation, and electrodeposition, and measured by alpha spectrometry using internal chemical yield tracers of ²⁴²Pu and ²⁴³Am. These methods are in extensive use in our studies (Shinn, 1993) of contamination sites around the world: Marshall Islands (Bikini, Enewetak, and Rongelap), Nevada Test Site (GMX, Little Feller, Palanquin, and Plutonium Valley), Tonopah-Nevada (Roller Coaster sites), Savannah River Lab-South Carolina (H Area),

Lawrence Livermore National Lab-California (Pu-garden site), Chernobyl-Ukraine (sandy beach site, near Nuclear Power Plant Unit 4), and Palomares-Spain (site of 1966 aircraft accident). Details of the protocol are provided in "Radiochemical Procedures for Analysis of Pu, Am, Cs, and Sr in Water, Soil, Sediments, and Biota Samples", (Wong et al, 1995).

Quality assurance was carried out through adherence to established protocols, (Kehl et al, 1995) and by use of quality control procedures (blank filters, control filters-carried to the site and back without use, and standards). Analytical accuracy has been maintained among these methods for the analysis of Pu (from atmospheric fallout) in shallow or deep marine sediments or in soil and sediment samples collected from the Pacific Test Sites at Bikini and Enewetak Atolls (close-in fallout). Our method has also been used in the analysis of Pu in NIST Radioactivity Standard Reference Materials (Rocky Flats Soil and Columbia River Sediment) and in IAEA interlaboratory comparison samples.

RESULTS

Activity Concentrations of Pu in Air

The median activity concentration of Pu in the suspended aerosols at the soil cleanup work site (combined SP, PA, and CP) was 3.64 pCi/g (135 Bq/Kg) in Oct-Nov 1992, as noted in the previous study (Shinn, Fry and Johnson, 1994). The changes we observed in subsequent monitoring of activity concentration probably reflects the changes in the activity of soil material processed. The median work-site activity concentration decreased to 0.43 pCi/g in Oct-Nov 1993, but increased to 57 pCi/g in the period Aug-Nov 1994, and to 38 pCi/g in Feb 1995. These median values are calculated from the data of Table 1, except for the 1992 case where the data are obtained from a larger data set reported by Shinn, Fry and Johnson, 1994. The estimate of the median in the cases of 1993, 1994, and 1995, was obtained by determining the geometric mean, for example the cube root of the product of three measurements at SP, PA, and CP.

The specific activity of suspended aerosols in the vicinity of the oversize soil crusher (OS) was always much less than in the other work areas. At OS the specific activity of suspended Pu was 0.14 pCi/g in Oct-Nov 1993, but increased to 2.5 pCi/g in Aug-Nov 1994, and to 7.7 pCi/g in Feb 1995; see Table 1. It was noted also that the specific activity of suspended Pu was slightly less near the clean pile (CP) than at the two work sites near the spoils pile and the plant area (SP and PA) except for 1992 when one air sample had unusually high values.

Table 1. Pu-Air Monitoring During Soil Sorting on Johnston Atoll, 1992-1995.

Monitoring location	Monitoring period	Pu Activity (pCi/g)	Pu in Air (aCi/m ³)	Mass Loading (µg/m ³)
Spoils Pile	14 Oct-14 Nov 1992	4.3	795	184
	20 Oct-15 Nov 1993	0.48	609	1280
	16 Aug-3 Nov 1994	81	45200	555
	17 Feb-27 Feb 1995	48	84200	1740
Plant Area	14 Oct-14 Nov 1992	3.4	322	94
	20 Oct-15 Nov 1993	0.44	105	238
	16 Aug-3 Nov 1994	93	42100	454
	17 Feb-27 Feb 1995	34	30600	888
Clean Pile	14 Oct-14 Nov 1992	9.2	689	82
	20 Oct-15 Nov 1993	0.37	73	199
	16 Aug-3 Nov 1994	24	6410	268
	17 Feb-27 Feb 1995	33	10300	315
Oversize Soil	14 Oct-14 Nov 1992	----	----	----
	20 Oct-15 Nov 1993	0.14	753	5340
	16 Aug-3 Nov 1994	2.5	1670	655
	17 Feb-27 Feb 1995	7.7	31800	4130
Background	14 Oct-14 Nov 1992	0.14	5.8	41
	20 Oct-15 Nov 1993	0.066	1.3	19
	16 Aug-3 Nov 1994	0.61	20.3	34
	17 Feb-27 Feb 1995	0.36	7.0	19

Mass Loading and Pu-Aerosol Concentrations

The total suspended particulate mass loading for the work area (SP, PA, and CP) increased significantly from a median of 109 µg/m³ in Oct-Nov 1992. Over the three monitoring periods in 1993 to 1995, the median TSP mass loading at SP was 1070 µg/m³, compared to 458 µg/m³ at PA and 256 µg/m³ at CP. But when the rock crusher was operating, the median total suspended particulates value at OS was 2440 µg/m³ and it was by far the more dusty site. By comparison, the upwind background HV measured a median 23 µg/m³ for the 1993-1995 period, and a significant portion of that mass loading would be due to the concentration of sea salt particulates. The Clean Air Act standard for TSP mass loading is 150 µg/m³ over a 24h period.

The Pu-concentrations of suspended particulates during the monitoring period varied both due to the amount of dust suspended in the work areas, and due to the activity of the soil. Multiplying the mass loading by the activity concentrations, we obtained the ^{239,240}Pu-aerosol concentration. See Figure 1.

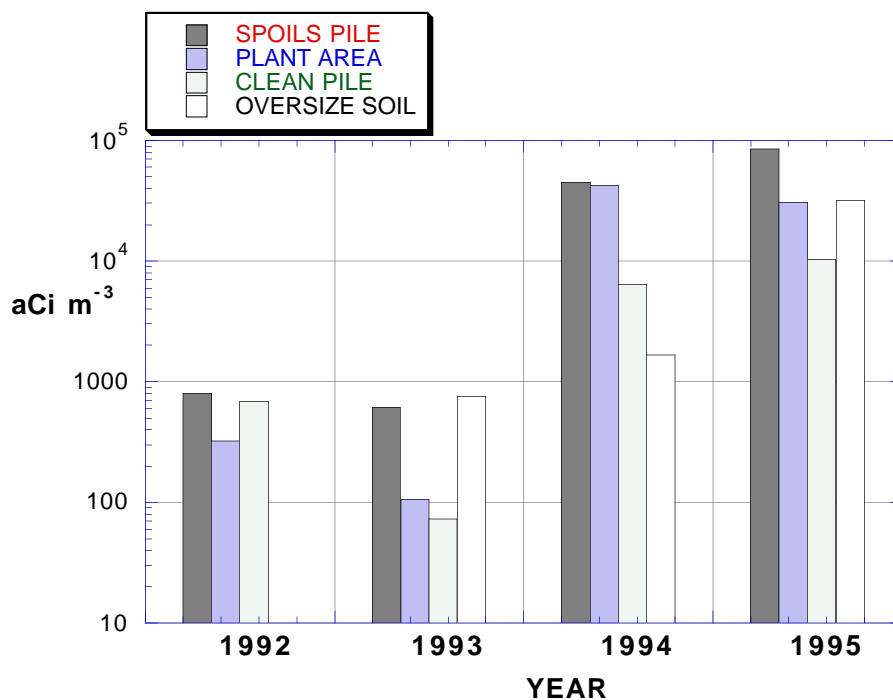


Figure 1. Pu in air (aCi/m³) during soil sorting operations at Johnston Atoll.

In 1992 the median concentration in the work sites (SP, PA, and CP) was 397 aCi/m³ (15 µBq/m³) for the period 14 Oct-14 Nov. The values of Pu in air ranged then from 280 to 1508 aCi/m³ (10 to 56 µBq/m³). The Pu concentration in air at the work sites decreased to median values of 168 aCi/m³ in Oct-Nov 1993, but increased to median values of 23000 aCi/m³ (852 µBq/m³) in Aug-Nov 1994 and 29800 aCi/m³ (1100 µBq/m³) in Feb 1995. (In the 1992 case the median is calculated from a larger data set reported by Shinn, Fry and Johnson, 1994. The estimate of the median in the cases of 1993, 1994, and 1995, was obtained by determining the geometric mean, for example the cube root of the product of three measurements at SP, PA, and CP.)

The spoils pile (SP) produced during 1993-1995 the highest Pu concentrations in air of all the work sites with a median Pu concentration of 13200 aCi/m³ (490 µBq/m³) and a range from 609 aCi/m³ to 84200 aCi/m³. The clean pile (CP) had the lowest median Pu concentration of 1690 aCi/m³ (63 µBq/m³). The

median value for the OS site near the rock crusher was 3420 aCi/m³ (127 µBq/m³) over the 1993-1995 period; higher mass loadings of total suspended particulates was counterbalanced by the lower activity of the particulates at the OS site. The rock crusher at OS site was operated 60% of the normal working hours during the test period, 1993-1995.

In 1992 the upwind, background mean ^{239,240}Pu-aerosol concentration was 5.8 aCi/m³ (0.21 µBq/m³). The median value for the background over the 1993-1995 period was 5.6 aCi/m³, with a range from 1.3 to 20 aCi/m³.

CONCLUSIONS

The work area certainly was a dusty environment, with median total suspended particulate levels that exceeded the Clean Air Act standard of 150 µg/m³ during most of the work hours, and reached median levels of 787 µg/m³ in 1995. The spoils pile area (median total suspended particulate mass loading of 1070 µg/m³) was more dusty than the plant area or the clean pile area. But the dust in the vicinity of the rock crusher at the oversize soil site was the worst with a median level of 2440 µg/m³ and a maximum of 5340 µg/m³ during the 1993-1995 observation periods.

Results of air monitoring for suspended Pu over the four periods in 1992-1995 during operation of a stationary soil sorting system on Johnston Atoll showed that there should be no high risk from worker exposure. Earlier studies (Shinn, Fry, and Johnson, 1994) showed that the median aerodynamic diameter of suspended mass and Pu-activity was 3.4 µm and would be in the middle of the "respirable" range. The concentration guide for radiation protection of workers exposed to ^{239,240}Pu (Lung Retention Class Y) is 6,000,000 aCi/m³ (2220 µBq/m³). This value is set by U.S. Department of Energy, in Federal Rule 10 CFR Part 835 (Federal Register, December 14, 1993.). The highest median value at the work sites (SP, PA, and CP) during the 1992-1995 monitoring period was 29800 aCi/m³ during Feb 1995, and this was more than 200 times lower than the regulatory level. The highest observed value was 84200 aCi/m³ at the spoils pile site, and this was more than 70 times lower than the regulatory level. Therefore, we do not find that the soil processing poses any significant risk to workers. See Table 2.

Table 2. Summary of Workplace Pu-Concentrations on J.A., 1992-1995.

Median Values in the Spoils Pile, Plant Area, and Clean Pile Work Area.

Monitoring location	Monitoring period	Pu in Air (aCi/m ³)	Mass Loading (μg/m ³)
Work Area (SP, PA, and CP)	14 Oct-14 Nov 1992	397	109
	20 Oct-15 Nov 1993	168	393
	16 Aug-3 Nov 1994	23000	407
	17 Feb-27 Feb 1995	29800	787
Background	1992-1995 median	5.7	27
Federal Standards		6,000,000	150

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REFERENCES

1. Bramlitt, E.T., 1993, Experience in Mining Plutonium for Soil Cleanup, p 27-32, in *Proceedings of the 1993 International Conference on Nuclear Waste Management and Environmental Restoration*, Prague, Czech Republic, Sept. 5-11, 1992, Vol. 2, eds. P.-E. Ahlstrom, C. C. Chapman, R. Kohout, and J. Marek, American Society of Mechanical Engineers, 345 47th St., United Engineering Center, New York, N.Y., 10017.
2. Shinn, J. H., C. O. Fry, and J. S. Johnson, 1994, *Suspended Plutonium Aerosols Near a Soil Cleanup Site on Johnston Atoll in 1992*, Technical Report UCRL-ID-116495, Lawrence Livermore National Laboratory.
3. Shinn, J. H., 1995, *Protocols of Radiocontaminant Air Monitoring for Inhalation Exposure Estimates*, Technical Report UCRL-ID-122254. Lawrence Livermore National Laboratory.
4. Shinn, J. H., 1993, *The Technical Basis for Air Pathway Assessment of Resuspended Radioactive Aerosols: LLNL Experiences at Seven Sites Around the World*, Technical Report UCRL-JC-115045, Lawrence Livermore National Laboratory, presented at Topical Workshop on the Technical Basis for Measuring, Modeling, and Mitigating Toxic Aerosols, Albuquerque, NM, Sept 27-30, 1993.
5. Wong, K. M, T. A. Jokela, and V. E. Noshkin, 1995, *Radiochemical Procedures for Analysis of Pu, Am, Cs, and Sr in Water, Soil, Sediments, and Biota Samples*, Technical Report UCRL-ID-116497, Lawrence Livermore National Laboratory.
6. Kehl, S. R., M. E. Mount, and W. L. Robison, 1995, *The Northern Marshall Islands Radiological Survey: A Quality Control Program for Radiochemical and Gamma Spectroscopy Analysis*, Technical Report UCRL-ID-120429, Lawrence Livermore National Laboratory.